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Polarization Studies in Fast-Ion Beam Spectroscopy

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In a historical review, the observations and the insight gained from polarization studies of fast ions interacting with solid targets are presented. These began with J. Macek's recognition of zero-field quantum beats in beam-foil spectroscopy as indicating alignment, and D.G. Ellis' density operator analysis that suggested the observability of orientation when using tilted foils. Lastly H. Winter's studies of the ion-beam surface interaction at grazing incidence yielded the means to produce a high degree of nuclear orientation in ion beams.

I. INTRODUCTION

For polarization to become observable, either the emitter must be restricted in its spatial orientation (for example, by a magnetic field), or the excitation must show directionality. Here I will give an overview of what in terms of polarization has been seen with (highly directional) fast ion beams that interact with targets in arrangements of various symmetries. Most of the work was done in the mid-1970s, but the topic has fallen dormant again after less than a decade. However, there are a few aspects of that work that are still of interest and that might even become useful again.

Since I have not worked in this particular sub-field myself (although I have worked with fast ion beams), I ask the reader to pardon me for my lack of depth and understanding, as well as for my more casual approach. Details of the formalism will be found, for example, in the contribution by A. Petrashen, and I am happy in not having to duplicate any of that excellent work. Instead of showing pictures from what is not my own work (which also would have required dealing with copyright issues), I shall show only a few sketches and refer to the literature for any details and specific presentations. The reference list is arranged alphabetically and includes the titles of the journal articles. This, I thought, might be more useful for anybody interested than me making up a serpentine presentation just in order to be then able to refer to this or that paper. Also, one finds quite a bit of repetition of the data and graphs presented in the original publications which are then re-hashed for this or that review or conference. Closer study might reveal specific information, if sought for, but I leave that to the interested reader. The list of references is far from complete. It was originally based on a US-based paper trail, and some European references have been added

from memory. For example, my own memory - backed up by recollections of colleagues - has traces of quantum beat work done at Lyon (France) in the late 1970s, by M. Ceyzeriat, A. Denis, and J. Désesquelles, but I have not found the references yet again. Looking up the beam-foil conference proceedings of the time will give further clues and references beyond this report.

I have structured my report along the following topics:

Historical context
Techniques and symmetries
Foil at right angles
- Coherent excitation
- Cylindrical symmetry $\rightarrow$ Alignment
- Best observed with a polarization analyzer
Quantum beats
- Fine structure intervals
- Hyperfine intervals
Tilted foil
- Cylindrical symmetry broken $\rightarrow$ Orientation
- Scattering at grazing incidence
(Ion-Beam Surface Interaction at Grazing Incidence - IBSIGI)
Dirt effects
- Temperature
- Current density
- Surface fields
- Surface contamination
Applications of IBSIGI
- Production of nuclear orientation for nuclear physics
- Probing the band structure of the surface material

II. HISTORICAL INTRODUCTION

At the beginning of the twentieth century, vacuum technology progressed to enable experiments with particle beams, be they electrons (for example, for producing x-rays) or ions (canal rays). Ion beam formation was being studied, both for investigations of the Stark effect and for analyzing the light intensity distribution along such canal rays. Wilhelm Wien realized that such a technique might reveal the lifetimes of atomic levels and derived approximate formulae for atomic level lifetimes long before quantum mechanics became available for that purpose [12].

In the 1930s, ion beams were adapted by nuclear scientists into accelerators (at the time branded/glorified as *atom smashers*) that could produce ions energetic enough to overcome the Coulomb barrier and induce nuclear reactions. Targets were now formed as thin foils which have a much higher areal density than a gaseous target. The ion beams would penetrate the target foil and then be analyzed for energy loss and charge state in a detector downstream, together with possible nuclear reaction fragments. The first ion beams were feeble, and people had difficulty in developing detectors for fast ions and reaction fragments,

and even detectors that would be sensitive enough to note and analyze a particle at all. With continuous development, ion sources improved and ion beams grew in beam current. However, it was only in the early 1960s that some nuclear physicists used their own eyes for detection again. They saw visible light from an ion beam after it had passed through a target foil. Several of them understood the atomic physics value of their observation [19,2] - some only to find themselves reprimanded by their faculty colleagues for straying away from nuclear physics [20].

Energetic ions passing through matter may lose one or more of their electrons. The remaining ones may be shaken into excited levels, or electrons may be captured upon leaving the target. In any case, this was a new way to produce multiply charged ions, populate high-lying or multiply-excited levels, and studying atomic processes with a time resolution of a small fraction of a nanosecond. This time-resolution was easily achieved in beam-foil spectroscopy, by mechanical means, displacing either the foil or the detector along the ion beam trajectory (in those days, tube-based electronics had a hard time in the ns time range). An ion beam of energy 2 MeV/nucleon travels about one centimeter in one nanosecond. Easily achieved mechanical displacements of 10 μm then correspond to one picosecond, without any need for fast electronics (and oscilloscope displays that were poorly suited for showing the traces of rapidly deflected electron beams). If one photographed the light emission that occurred along the ion beam (using a filter to discriminate one spectral line from the others), the change of light intensity was interpreted as an intensity decay curve. The time after excitation strictly correlates with the distance from the foil via the uniform ion beam velocity. After properly converting film blackening back to an intensity scale, one might then determine atomic decay time constants. Photoelectric detectors were rather novel then, but quickly took over.

However, while there were many promises of the new technique [3], there were also many puzzling features that required some time to be recognized and sorted out. For example, some of the intensity decay curves, in particular those from hydrogen atoms or H-like ions, showed oscillations. These were quickly subsumed as resulting from "something like the Hanle effect", invoking stray electric and magnetic fields, or even fields produced by the ion beam itself, as well as the degeneracy of levels in hydrogenlike ions. In 1969, however, Joe Macek (then at Lincoln, Nebraska) suggested that the oscillations might have quite another origin. This origin is what we now call coherent excitation, and would lead to zero-field quantum beats, which are best observed with polarization-sensitive equipment [23]. This concept brings the story to the topic of this polarization spectroscopy workshop. A little more detail is needed to explain the how and why.

III. FOIL AT RIGHT ANGLES

The ion beam trajectory provides a direction, and any sideways orientation in a plane whose normal coincides with the beam direction has no preference over any other. The standard arrangement of an exciter foil in beam-foil spectroscopy is just that, with a foil at right angles, i.e., the foil surface normal pointing along the ion beam (Fig. 1).

Such a thin foil (with an areal density of, say, 10 to 20 μg/cm^2, or about 1/500 of the thickness of writing paper) is being traversed by the ion beam in a time of order 10^{-14} s. This time interval is comparable to the typical lifetimes of K-shell vacancies in (moderately) heavy ions. However, for most of the subsequent discussion, I shall concentrate on visible

light, from the excitation of valence electrons in not so highly charged ions. There the typical lifetimes range from 10^{-9} s to 10^{-11} s, which is orders of magnitude longer than the time the ions spend inside the foil. Inside the bulk of the foil, electronic excitation of the fast ions is very likely, but the cross sections for electron loss are also very high, and excited states are easily destroyed again before the ion leaves this dense environment. Excitation in the surface layer may survive, and more population of excited levels may occur by electron capture after leaving the foil. Therefore the typical excitation has to happen in a time interval that is even shorter than the time it takes to cross the foil, and this puts the time interval close to 10^{-15} s.

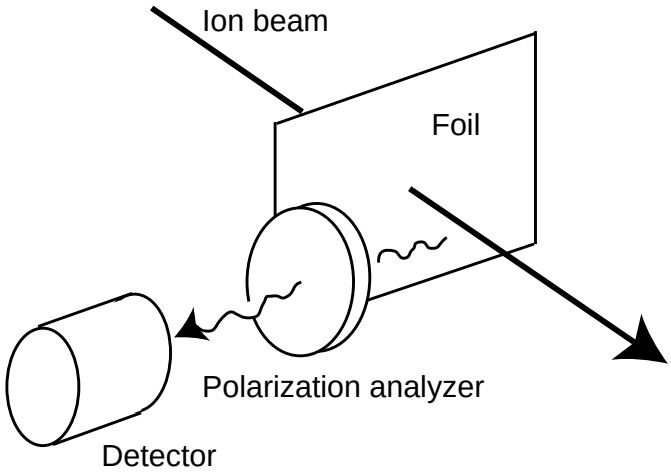

FIG. 1. Geometry with a fast-ion beam and an exciter foil at right angles.

Such considerations paved the ground for a conjecture by Macek [23] who suggested that the oscillations observed on decay curves might relate not to external fields, but be the consequence of coherent excitation. In this picture, via the uncertainty principle, a short time interval corresponds to an uncertainty in excitation energy. Excited levels with an energy separation smaller than this energy spread would be coherently excited, and their decay would possibly display interference effects.

Take two fine structure levels of a given term as an example and consider their decays to another level. As with coupled pendulums in mechanics, the levels develop in time together, and the level energy difference ΔE shows as a frequency ν in the decay curve. As the frequency describes a (lower-frequency) beat pattern between two high frequencies (the actual transition energies) associated with transitions between quantum states (and does not require external fields), the phenomenon has been dubbed *zero-field quantum beats*. In a simple view, this phenomenon results from the non-statistical population (different excitation cross sections) of the magnetic sublevels m_l effected in the ion-foil interaction. With a foil at right angles to the ion beam, the only asymmetry expected may be forward/backward, and this restricts the dependence of the sublevel population effect to a dependence on the absolute values of m_l, $|m_l|$.

Such *alignment* can then only affect terms with levels of $|m_l| > 1/2$. It is thought of to result (semiclassically) in a rotating dipole that is best observed by a polarization analyzer. At a certain *magic angle* (of 54.7°, where the Legendre polynomial P_2 vanishes), it is possible to obtain a signal to which all polarization components contribute in a way that renders the result independent of alignment and which thus is proportional to the total intensity of the decay (the total intensity shows no beat pattern). If one wants to measure the alignment, one measures through a polarization analyzer at two mutually perpendicular orientations (for example, along/across the ion beam) the signal strength and then applies a density operator analysis or uses the Stokes parameters (outlined in Table III). Remember that a J=1/2 - 1/2 transition cannot show polarization (which makes it a useful unpolarized sample), but the 1/2 - 3/2 component can. So in atomic lifetime measurements employing either a fast ion beam and a foil, or short-pulse laser excitation of atoms in a vapor cell, one tries to obtain unperturbed decay curves and thus avoids quantum beats. On the other hand, searching for level population effects or trying to measure small term differences in an otherwise poorly accessible range of level energies, it is advantageous to seek quantum beats.

TABLE I. The Stokes Parameters help in evaluating the measured polarization signal in terms of the density matrix, illuminating the physical situation.

$I = \|E_{\|}\|^2 + \|E_{\perp}\|^2$
$M = \|E_{\|}\|^2 - \|E_{\perp}\|^2$
$C = 2\ \mathrm{Re}\ (E_{\|}E_{\perp}^*)$
$S = 2\ \mathrm{Im}\ (E_{\|}E_{\perp}^*)$
With I being the overall intensity,
$M/I = \langle\ L_y^2 - L_z^2\ \rangle\ /\ \langle\ L_x^2\ \rangle$ relates to the alignment,
$C/I = 2\ \mathrm{Re}\ \langle\ L_y L_z\ \rangle\ /\ \langle\ L_x^2\ \rangle$ measures the correlation of L_y and L_z, and
$S/I = -\ \bar{h}\ \langle\ L_x\ \rangle\ /\ \langle\ L_x^2\ \rangle$ relates to circular polarization.

An experimental problem with quantum beats is their frequency. A decay curve of a level with a multi-nanosecond lifetime may stretch out over a few centimeters along the ion beam. In order to analyze the beat pattern, the frequency has to be high enough to result in many oscillations. Yet the frequency must be low enough to be resolved spatially (which, as mentioned above, corresponds to time resolution). This limits observations largely to low charge states and to ions with a small fine structure, that is with a single electron in the valence shell. For early examples, see the work done in Andrä's group in Berlin and in Bukow's group at Bochum [1,15,33,30]. However, there also are ions in which hyperfine intervals can be seen from quantum beats, and then often quite a number of hyperfine level intervals can be recovered from a Fourier analysis of the beat pattern.

Measurements of alignment do not necessarily require the observation of quantum beats, only of polarization. Still, polarizers with sufficient analyzing power and transmission largely restrict such work to wavelengths in the visible spectrum. A remarkable exception is the Ly_α transition in hydrogen, with a wavelength near 121.5 nm. Several polarizers of different designs have been built for this wavelength, using, for example, a stack of tilted LiF plates (in transmission) or a set of three surface reflections [30]. Much of this work was done by Helmut Winter (Bochum/Lyon/Berlin/Münster/Berlin). He wanted to ascertain the ion-energy variation of the alignment (energy dependence of the relative cross sections of the magnetic sublevels) in order to find the predicted cross-over between positive and negative values of the alignment. He did find a first crossing for about 50 keV H atoms [30], and later on an even lower-energy second crossing. By then he interpreted his data no longer as an energy trend, but, theoretically justified, as a variation with the inverse of the ion velocity [21]. In order to remind the plasma workshop audience of the energy scale: 50 keV protons colliding with the electrons in the foil correspond to 25 eV electrons impinging on protons in a plasma.

IV. TILTED FOIL

Macek's article referred to a much earlier treatment of polarization, by Percival and Seaton [24], who had considered the excitation of atoms by an electron beam, extending the even earlier Oppenheimer-Penney approximation. In 1973, Dave Ellis from Toledo (Ohio) revisited the case and expanded the scope by breaking the dominant symmetry [16]. By tilting the exciter foil (Fig. 2), he suggested, the previously used cylindrical symmetry around the ion beam would be broken, and Percival and Seaton's formalism would lead from the density matrix to a tensor with a main axis tilted away from the ion beam. In this way, *orientation* would be added to the alignment, to be observed as a superposition of linear and circular polarization, resulting in elliptical polarization with a major axis of specific orientation in space. The underlying level population would then no longer depend just on absolute values $|m_l|$, but on m_l itself.

The University of Toledo had an ion accelerator, and the theoretician Dave Ellis and his more experimentally inclined colleagues Gordon Berry, Larry Curtis and Dick Schectman promptly confirmed all these predictions. They then continued on to explore the parameter space, devising 3-D contour plots to follow the Stokes parameters as a function of ion energy and foil tilt angle. Via the coupling of nuclear and electron-shell momenta, it could be shown that the orientation of the electron shell could be transferred to the nucleus, and a beam of oriented nitrogen nuclei could be obtained [7].

One of the first surprises was that the orientation grew with the tilt angle to very high values (then the projection of the foil in the direction of the ion beam increases the ion path through the foil so much that the ions are lost due to energy loss and scattering). This contradicted an earlier model that had assumed excitation to take place mostly inside the foil, and clearly favored a dominance of surface effects. In fact, the observed orientation simply corresponded to what one would expect from the relative motion of ions outside a tilted surface, picking up largely stationary electrons, or from a surface friction picture. In Fig. 2, the observer/detector would see right-hand circular polarization.

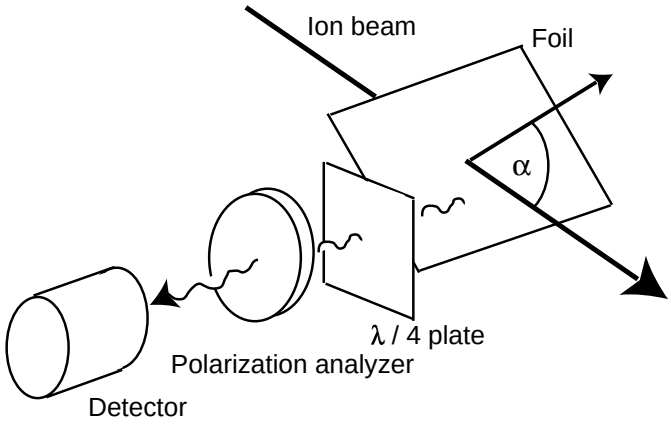

FIG. 2. Geometry with a fast-ion beam and an exciter foil that is tilted from the ion beam by an angle α. Since some of the observations seek for circular polarization, a $\lambda/4$ plate is required to convert this part of the light into linearly polarized light first.

Of course, such pictures must be oversimplified, and thus some rather fierce discussion broke loose, each side having evidence (from perpendicular or tilted foils) supporting their own views and contradicting others. One study found a dependence on the temperature of the foil (that under intense ion bombardment can glow brightly) [17]. Another study claimed this to be a current-density effect instead [29]. In their model, the limited conductivity of carbon foils trying to replenish secondary electrons released by ion bombardment would create a current in the foil, converging onto the ion beam spot, with associated electric and/or magnetic fields. This current-density effect might be influenced by the choice of foil material, special backings, foil holders or supporting grids. A third group found no current-density effect even when varying the current density in a wider range than the second had done [22]. The foil material was varied [11], then finding that carbon and similar non-metals yielded more polarization than metals did. Soon it was found that the ubiquitous contamination of materials under ion bombardment by cracked hydrocarbons of whatever origin clouded the sensitivity of the experiments. And so on.

On the theoretical side, the corrugation of a foil surface, the state density in and above the surface, various intricate models for electron capture, the influence of surface fields, and whatever else were invoked - and made no recognizable impact on the results. However,

a few insights survived, supported by evidence from other experiments: Inner vacancies are mostly produced in the bulk of the foil material, whereas much of the valence electron structure is assembled only in the surface layers of the foil, if not above the surface. Surface fields play a notable role only for hydrogenic (degenerate or near-degenerate) levels.

Orientation was found in multiply charged ions [18], too. However, there was no valid model to describe the various polarization effects of outer electrons theoretically, and for lack of that and for exhaustion of accessible simple atomic systems, the field of polarization studies in beam-foil spectroscopy petered out.

V. SCATTERING AT GRAZING INCIDENCE

Some of the above can be driven to the extreme, and there gain new significance: At very large tilt angles, the ions can no longer penetrate the foil. Instead, one may try to scatter them from the same surface, under grazing-incidence conditions. This then almost implies a surface without a bulk; it also requires ultra-high vacuum and in-situ cleaning of the surfaces by ion sputtering. Again, Helmut Winter tried his hands on this, and again successfully. His IBSIGI technique (Ion-Beam Surface Interaction at Grazing Incidence), begun in collaboration with H.J. Andrä, provided unprecedentedly high degrees of orientation, including a high-yield arrangement for producing a beam of nitrogen ions with nuclear spin polarization [31]. The nuclear spin polarization reached 15% after scattering on one surface, and 20% after scattering off two such surfaces. The technique has also been employed to study surface fields between an incoming ion (with the vertical velocity component reduced by geometry) and the target surface, and it has recently been used to probe the electron band structure of a composite material inside the target bulk. On the fundamental physics side, Winter has demonstrated with individual ion-atom collisions that the aforementioned simple geometrical picture of producing orientation holds. The latest feat of Winter's group is a measurement of the circular polarization of Auger electrons after IBSIGI on magnetized surfaces [25]. If you want more details, look for the extensive Physics Report that he has just finished preparing.

VI. CONCLUSION

Why is polarization spectroscopy of fast ion beams a (largely) closed chapter? Probably because there is no conclusive theory predicting the polarization of specific outer electrons.

Inner-shell vacancies in heavy ions, produced by ion-foil interactions, have occasionally been studied by evaluating angular correlations of the x-ray emission pattern, but hardly by employing polarization-sensitive devices. This may become an interesting field (again), because plasma physics has diagnostic problems at very-high density. In such plasmas, collisions are so frequent that multiple excitation becomes possible - a mainstay of beam-foil spectroscopy for almost four decades. Beam-foil spectroscopy features ion excitation in a dense medium (solid matter), followed by observation in a low-density medium (good vacuum). There may be prospects ...

Acknowledgment: Larry Curtis and his web-based paper mine, with access to his many insightful manuscripts, made it easy to dig into this field. Let me thank Dave Ellis and

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